LA-UR-17-30269

Title:	Nuclear Forensics and Radiochemistry: Radiation Detection
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Intended for:	UC Berkeley lecture series.
Issued:	2017-11-08

Nuclear Forensics and Radiochemistry: Radiation Detection

Robert S. Rundberg

Abstract: Radiation detection is necessary for isotope identification and assay in nuclear forensic applications. The principles of operation of gas proportional counters, scintillation counters, germanium and silicon semiconductor counters will be presented. Methods for calibration and potential pitfalls in isotope quantification will be described.

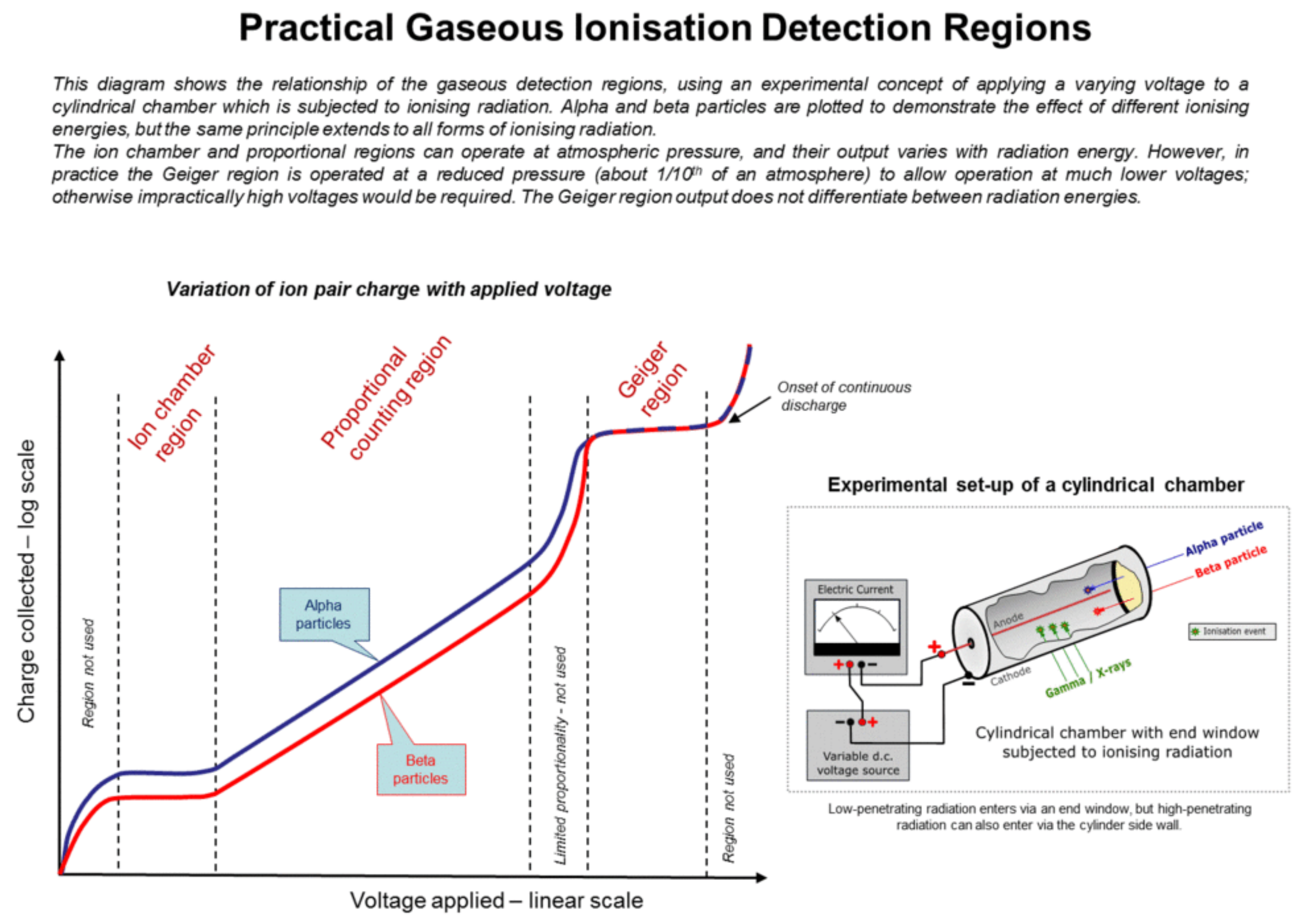
Nuclear Forensics and Radiochemisty: Radiation Detection

Lecture3

Gas Proportional Counters and Ionization Chambers

- Los Alamos fission product yields rely heavily on beta counting with gas proportional counters.
  - This requires chemical separation from interfering isotopes because the beta spectrum is a continuum.
  - The robust separation methods used at Los Alamos are documented in LA-1721
  - The advantage of beta counting is that fission products being neutron rich decay by beta emission the efficiency is generally high ~30 percent. A beta is emitted with every decay, whereas gamma counting may require additional information to obtain a decay rate (branching ratios, internal conversion coefficients, etc.)
  - Samples must be prepared in a reproducible manner. The size of the deposit is always the same. The count rate must be corrected for the thickness of the deposit.
- Gas proportional Counters still have particle energy resolution, while electron multiplication gives sensitivity to low energy betas.
- The Fano factor is about 0.2.

The Proportional Counting Region



Practical Gaseous Ionisation Detection Regions

This diagram shows the relationship of the gaseous detection regions, using an experimental concept of applying a varying voltage to a cylindrical chamber which is subjected to ionising radiation. Alpha and beta particles are plotted to demonstrate the effect of different ionising energies, but the same principle extends to all forms of ionising radiation.

The ion chamber and proportional regions can operate at atmospheric pressure, and their output varies with radiation energy. However, in practice the Geiger region is operated at a reduced pressure (about 1/10th of an atmosphere) to allow operation at much lower voltages; otherwise impractically high voltages would be required. The Geiger region output does not differentiate between radiation energies.

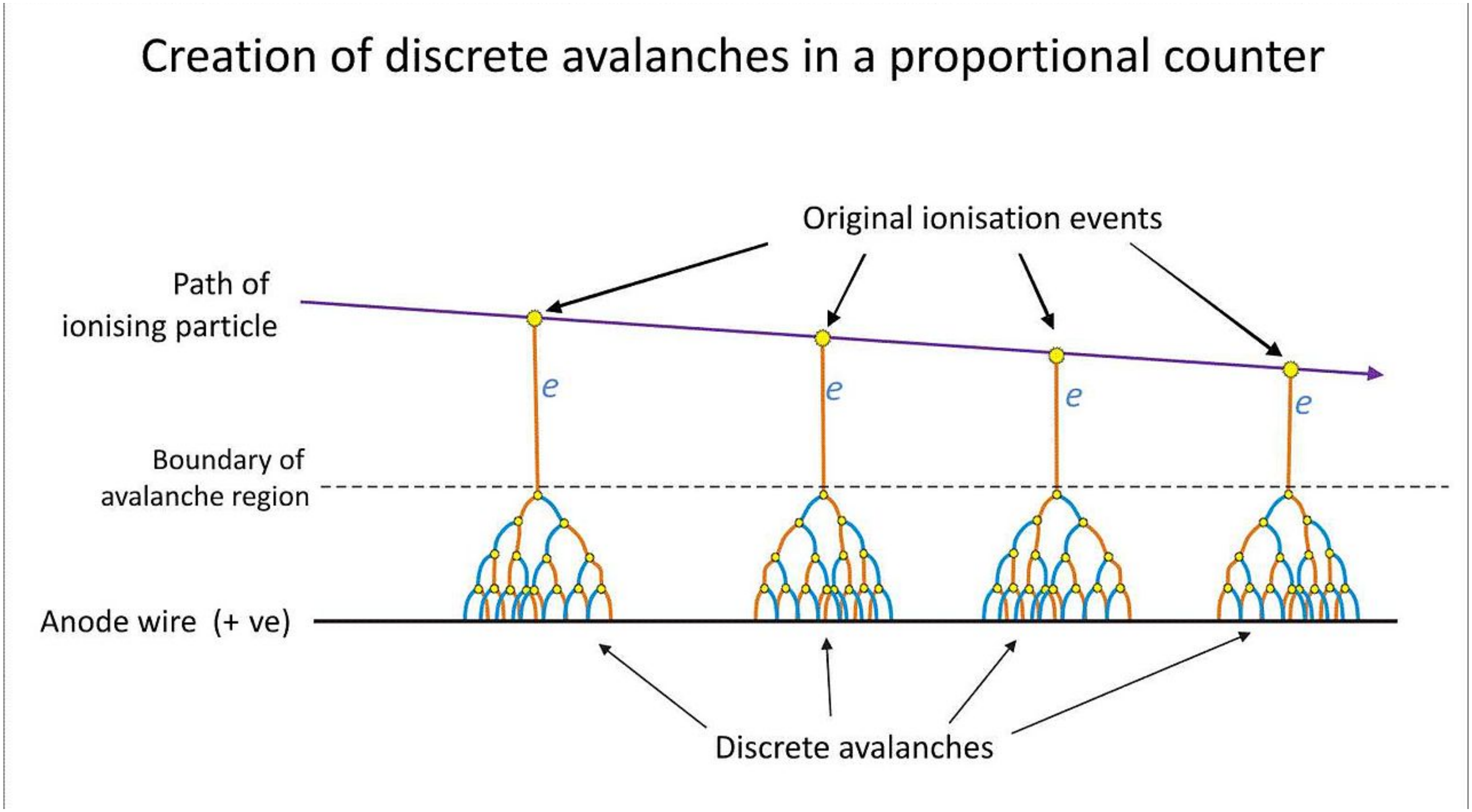
Variation of ion pair charge with applied voltage

Experimental set-up of a cylindrical chamber

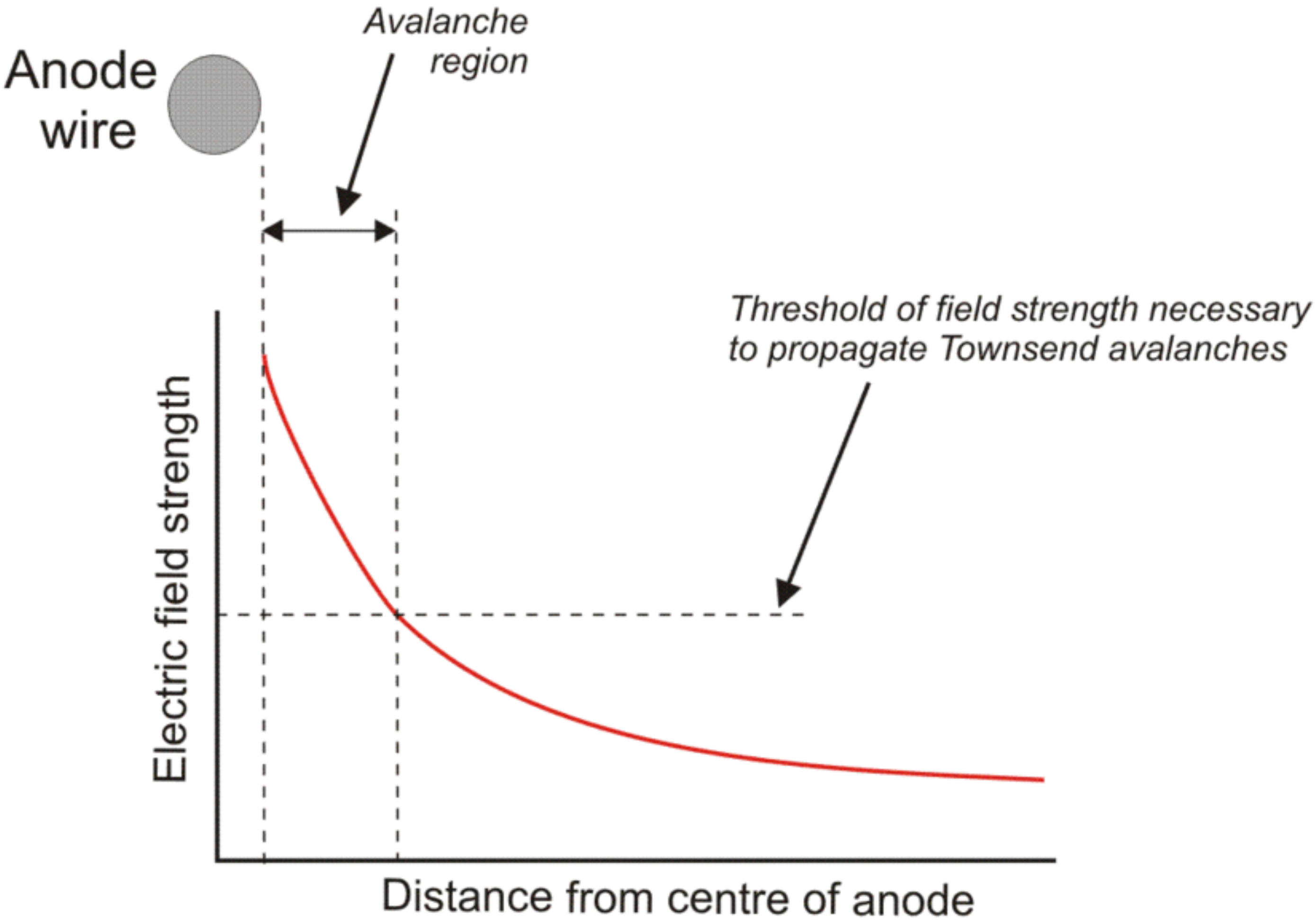
Cylindrical chamber with end window subjected to ionising radiation

Low-penetrating radiation enters via an end window, but high-penetrating radiation can also enter via the cylinder side wall.

How Charge Multiplication is Accomplished



Electric field strength at a counter anode



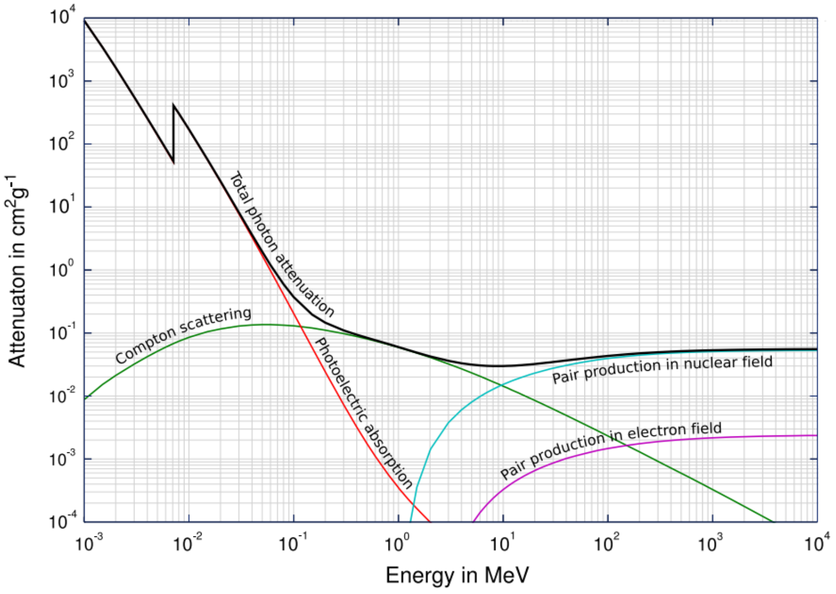
Equation for Multiplication

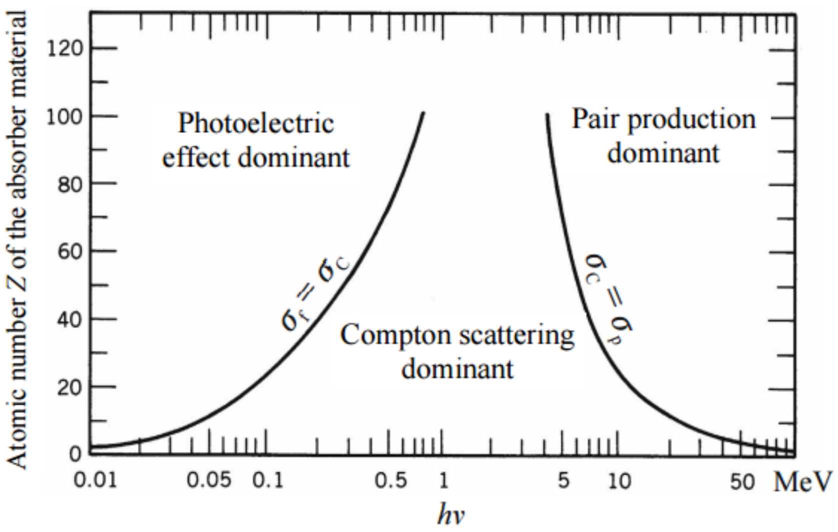
The equation for multiplication, i.e., gain, in a gas proportional counter

$$\ln M = \frac{V}{\ln(a/b)}\frac{\ln 2}{\Delta V_\lambda}\left[\ln\left(\frac{V}{pa\ln(b/a)}\right) - \ln K\right] \tag{1}$$

Where a is the anode wire radius, b is the radius of the counter, p is the pressure of the gas, and V is the operating voltage. K is a property of the gas used and relates the energy needed to cause an avalanche to the pressure of the gas. The final term ΔV_λ gives the change in voltage caused by an avalanche.

Detectors and the Interaction of Gamma Rays with Matter



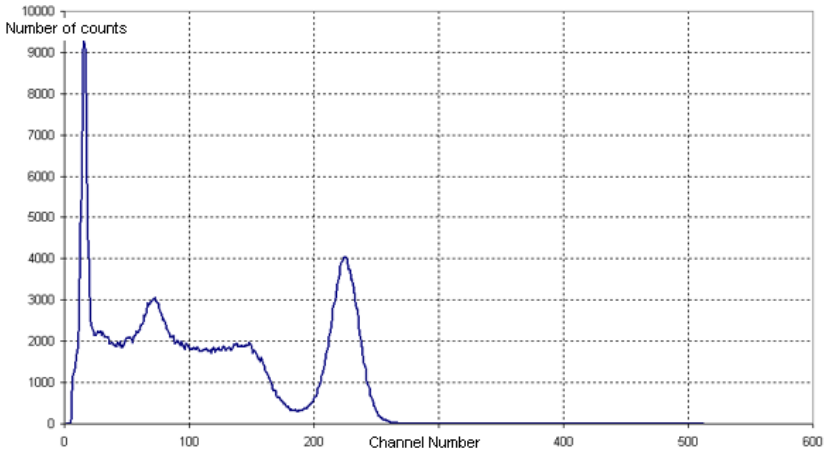


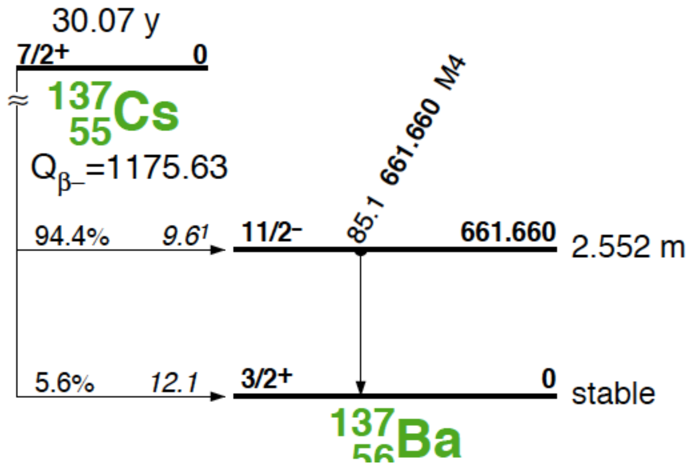
- The photoelectric effect is most likely to result in a photopeak.
- The higher the atomic number of the detector material the better.

The Physics of Scintillators

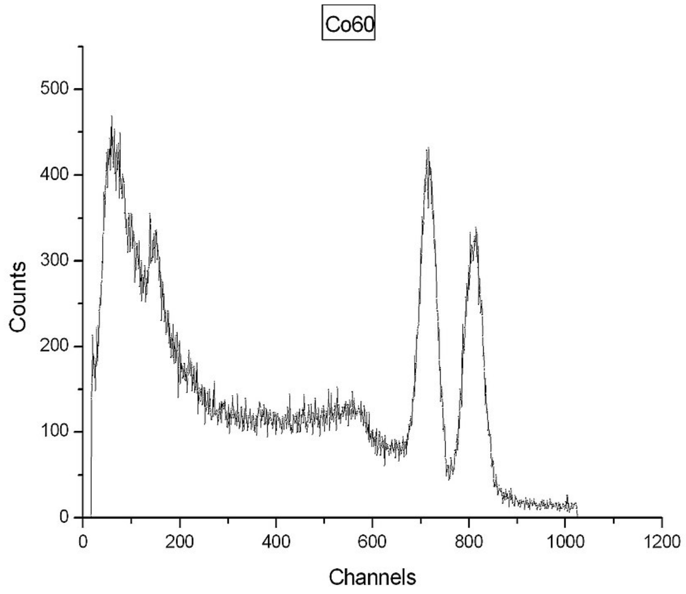
The scintillation process in inorganic materials is due to the electronic band structure found in crystals and is not molecular in nature as is the case with organic scintillators.[17] An incoming particle can excite an electron from the valence band to either the conduction band or the exciton band (located just below the conduction band and separated from the valence band by an energy gap; see picture). This leaves an associated hole behind, in the valence band. Impurities create electronic levels in the forbidden gap. The excitons are loosely bound electron-hole pairs which wander through the crystal lattice until they are captured as a whole by impurity centers. The latter then rapidly de-excite by emitting scintillation light (fast component). The activator impurities are typically chosen so that the emitted light is in the visible range or near-UV where photomultipliers are effective. The holes associated with electrons in the conduction band are independent from the latter. Those holes and electrons are captured successively by impurity centers exciting certain metastable states not accessible to the excitons. The delayed de-excitation of those metastable impurity states again results in scintillation light (slow component).

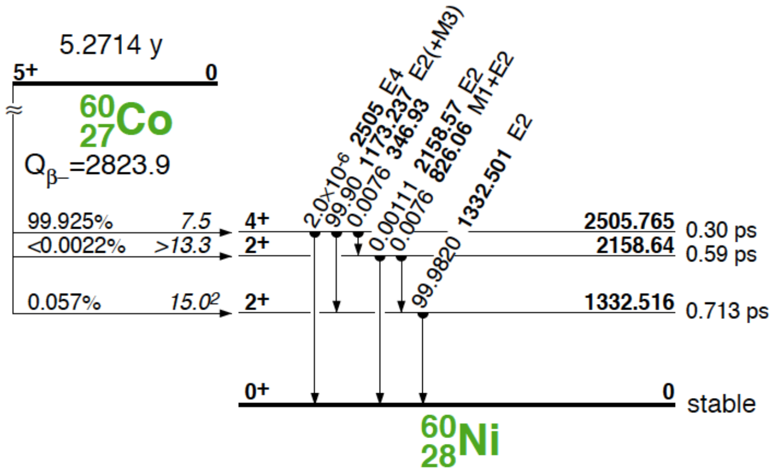
NaI(Tl) Spectrum of Cs-137





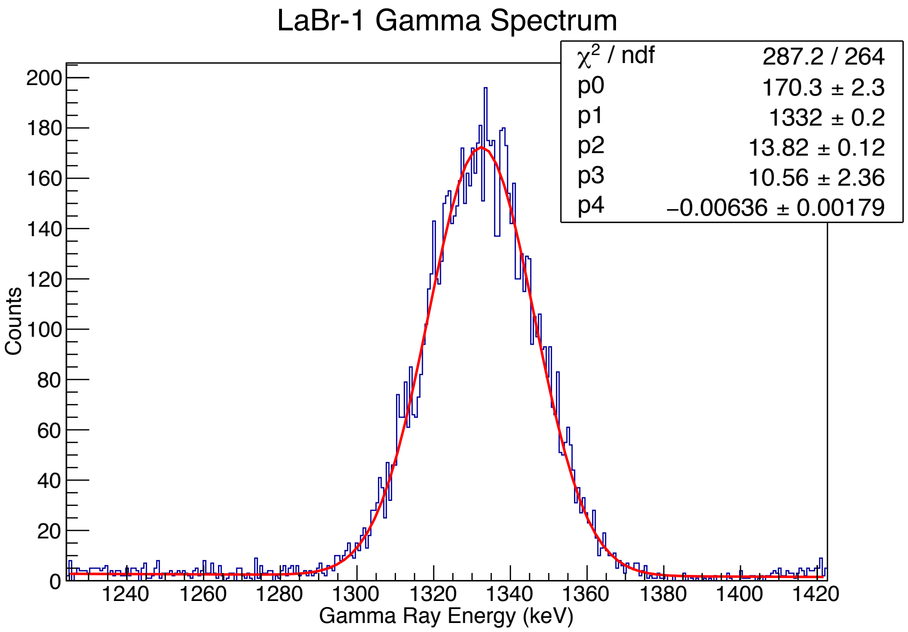
NaI(Tl) Spectrum of Co-60



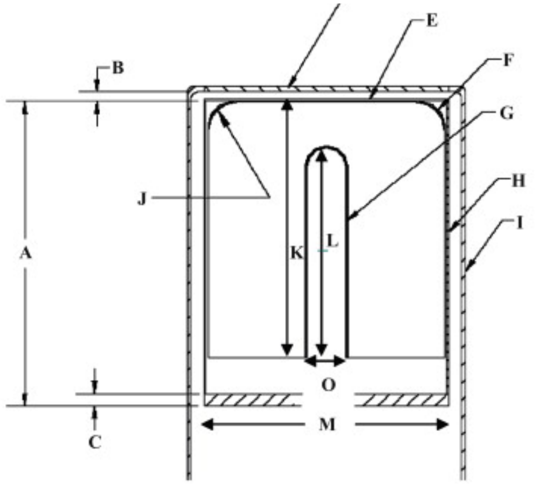


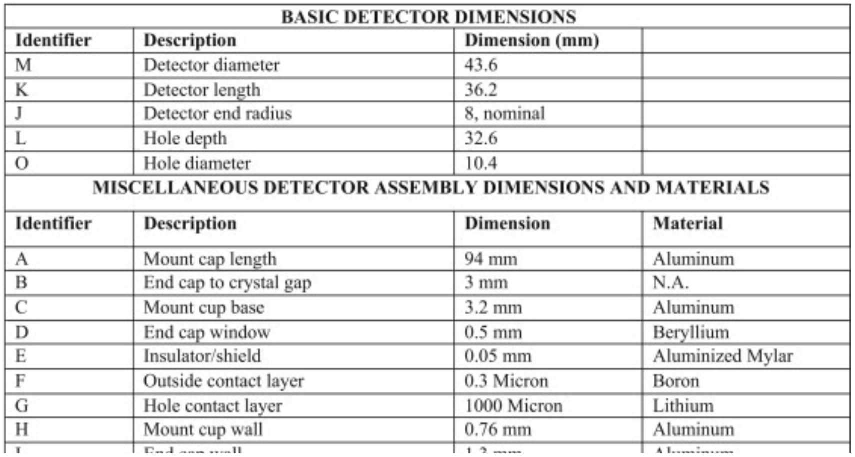
The Energy Resolution of Scintillators

- Scintillators are fit well by a Gaussian.
- Scintillators should obey Poisson Statistics, i.e., the expected number of events equals the variance.
- LaBr(Ce) yields 63 photon per keV. 1332 keV = 8.39E4 photons.
- The quantum efficiency of a typical photo-cathode is 20%.
- Sigma is therefore 129 e^- or 10.2 keV.
- The Fano factor is 1.0.



High Resolution HPGe Detectors



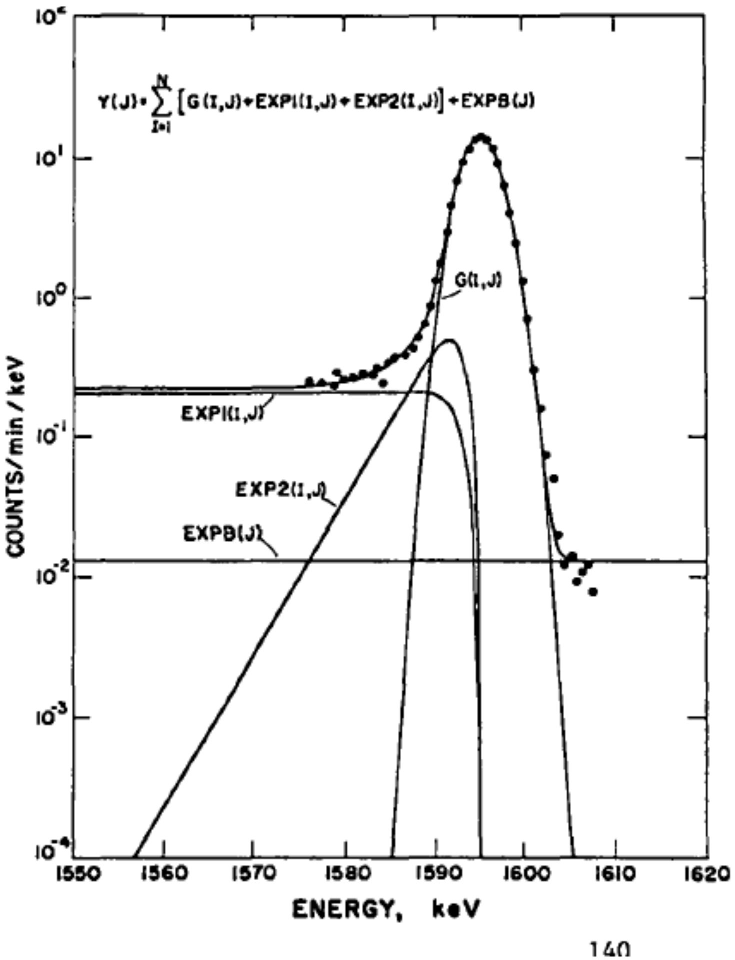


BASIC DETECTOR DIMENSIONS			
Identifier	**Description**	**Dimension (mm)**	
M	Detector diameter	43.6	
K	Detector length	36.2	
J	Detector end radius	8, nominal	
L	Hole depth	32.6	
O	Hole diameter	10.4	

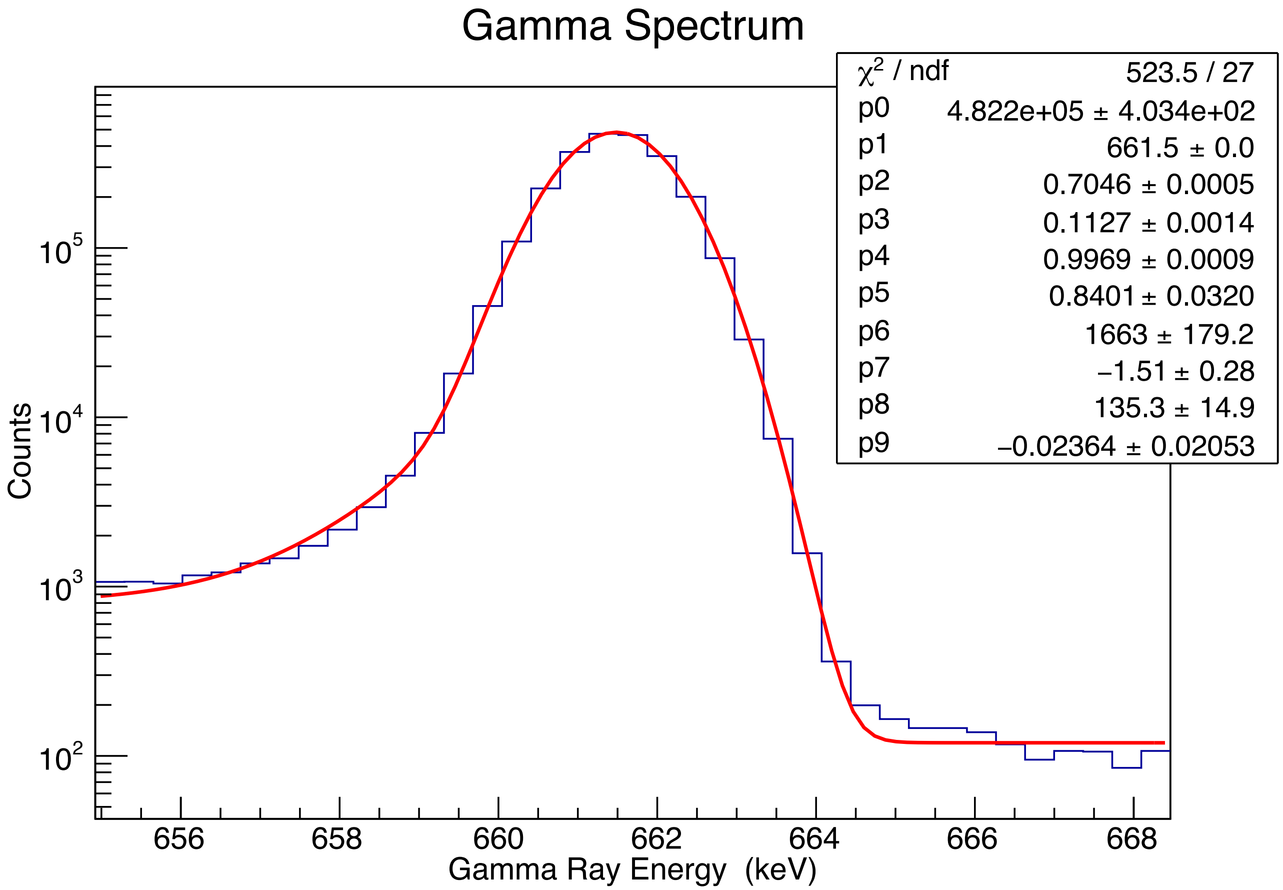
MISCELLANEOUS DETECTOR ASSEMBLY DIMENSIONS AND MATERIALS			
Identifier	**Description**	**Dimension**	**Material**
A	Mount cap length	94 mm	Aluminum
B	End cap to crystal gap	3 mm	N.A.
C	Mount cup base	3.2 mm	Aluminum
D	End cap window	0.5 mm	Beryllium
E	Insulator/shield	0.05 mm	Aluminized Mylar
F	Outside contact layer	0.3 Micron	Boron
G	Hole contact layer	1000 Micron	Lithium
H	Mount cup wall	0.76 mm	Aluminum
I	End cap wall	1.3 mm	Aluminum

The Shape of an HPGe Photopeak

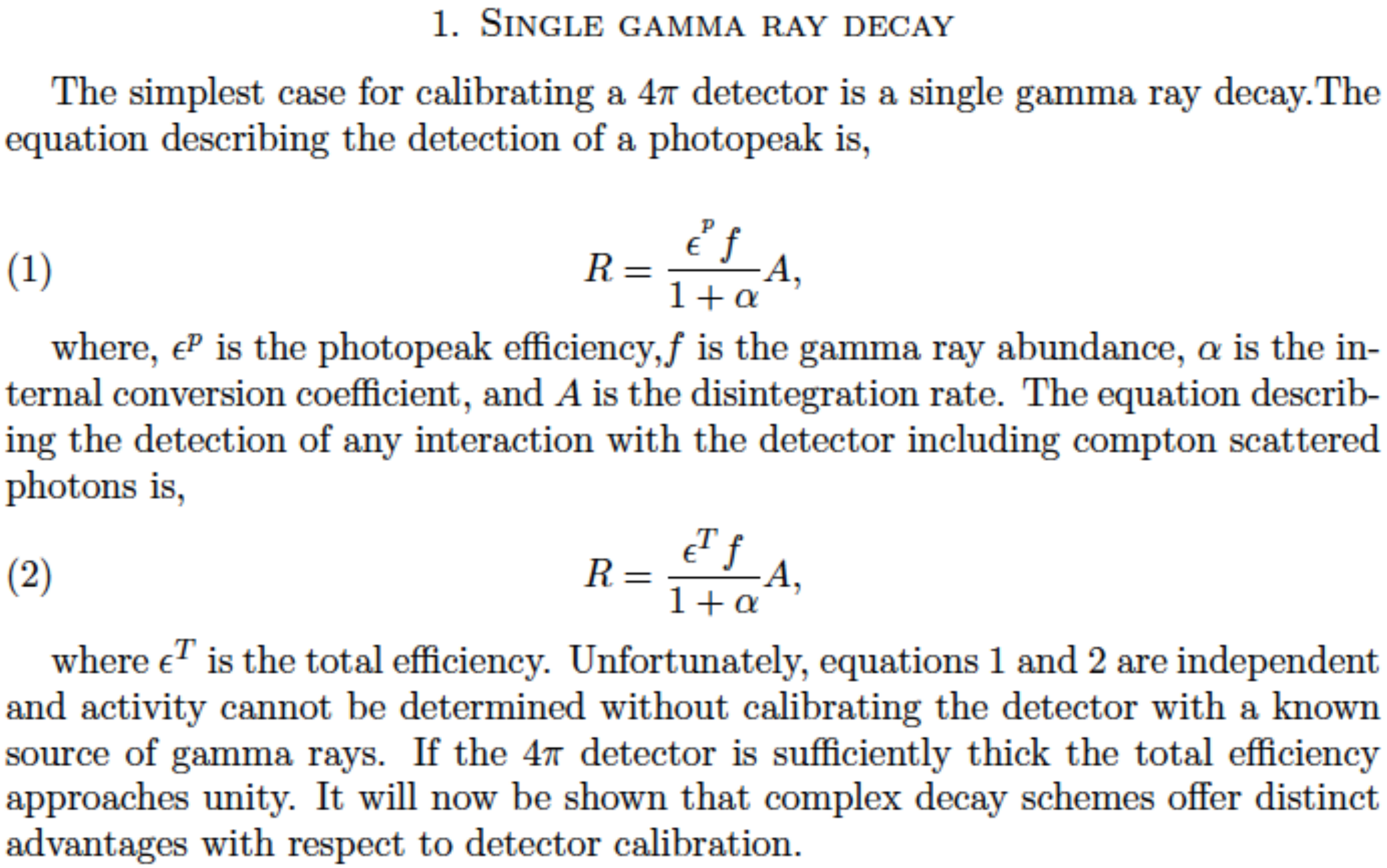
- Sanders and Holm, LA-4030, (1969).
- Consists of:
  - A Gaussian
  - An exponential tail, terminated by multiplying by 1 minus the gaussian (due to charge trapping)
  - A zero slope tail also , terminated by multiplying by 1 minus the gaussian (due to Rayleigh scattering of photons)
  - A zero slope background (while appropriate for this La-140 line should be generalized to a polynomial).



Fit of C-137 Gamma Spectrum



Gamma Ray Calorimetry



1. SINGLE GAMMA RAY DECAY

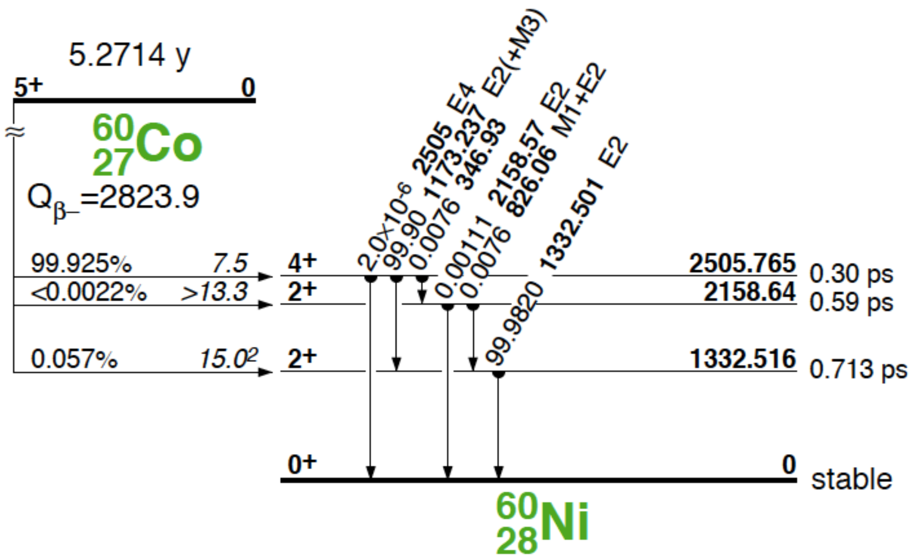
The simplest case for calibrating a 4π detector is a single gamma ray decay.The equation describing the detection of a photopeak is,

$$R = \frac{\epsilon^{p} f}{1+\alpha} A, \tag{1}$$

where, ϵ^p is the photopeak efficiency,f is the gamma ray abundance, α is the internal conversion coefficient, and A is the disintegration rate. The equation describing the detection of any interaction with the detector including compton scattered photons is,

$$R = \frac{\epsilon^{T} f}{1+\alpha} A, \tag{2}$$

where ϵ^T is the total efficiency. Unfortunately, equations 1 and 2 are independent and activity cannot be determined without calibrating the detector with a known source of gamma rays. If the 4π detector is sufficiently thick the total efficiency approaches unity. It will now be shown that complex decay schemes offer distinct advantages with respect to detector calibration.



2. Two level gamma ray cascades

The problem with calibrating a 4π detector with a two level decay scheme is summing of part or all of the energy from both gamma rays. If the two gamma rays are in coincidence the observed rate for the photopeak of gamma ray 1 is:

$$R_1 = \frac{\epsilon_1^p f_1}{1+\alpha_1}\left(1 - \frac{\epsilon_2^T f_2}{1+\alpha_2}\right)A = \xi_1^p(1-\xi_2^T)A \tag{3}$$

where, ϵ_1^p is the photopeak efficiency for gamma ray 1, ϵ_2^T is the total efficiency (Comptons and photopeak) for gamma ray 2, and A is the disintegration rate. Likewise the observed rate for the photopeak of gamma ray 2 is:

$$R_2 = \xi_2^p(1-\xi_1^T)A \tag{4}$$

The sum peak is observed when the full energy of both gamma rays are absorbed. The sum peak rate is:

$$R_{12} = \xi_1^p \xi_2^p A \tag{5}$$

The fourth observable is the rate of Comptons plus photopeaks. This rate is

$$R_{tot} = \xi_1^T A + \xi_2^T A - \xi_1^T \xi_2^T A \tag{6}$$

The ratio of singles to sum peaks is proportional to the activity,

$$\frac{R_1 R_2}{R_{12}} = (1 - \xi_1^T)(1 - \xi_2^T)A = A - \xi_1^T A - \xi_2^T A + \xi_1^T \xi_2^T A$$

thus, the activity is,

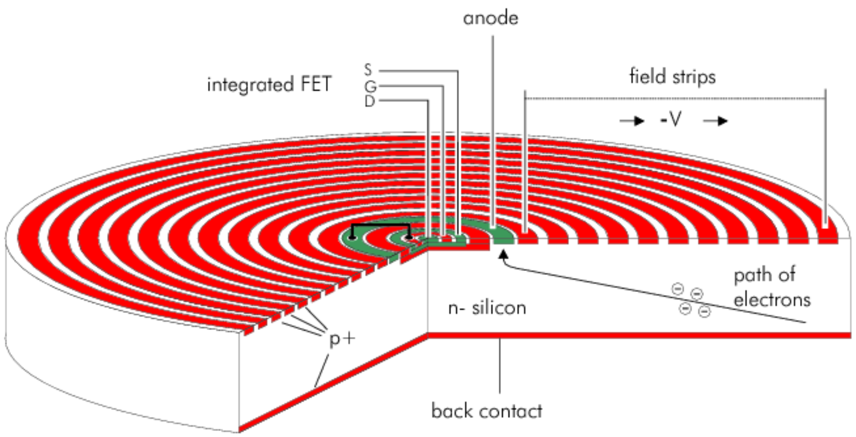
$$A = \frac{R_1 R_2}{R_{12}} + R_{tot}, \tag{7}$$

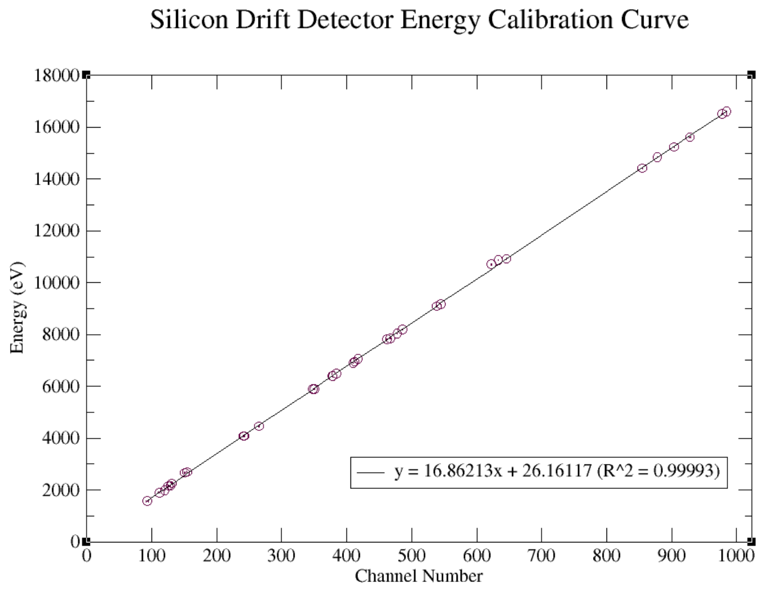
independent of the detector efficiency. The first term in equation 5 becomes small as the 4-Pi detector efficiency becomes large.

Gamma Ray Calorimetry

- Absolute disintegration rate can be determined without knowing the efficiency.
- Applies only to decay branches that have gamma rays or detectable x-rays.
- In the past, at Los Alamos, sources were counted in 3x3, 5x5, and 11x11 NaI(Tl) well detectors. The approach towards 100% efficiency was observed.

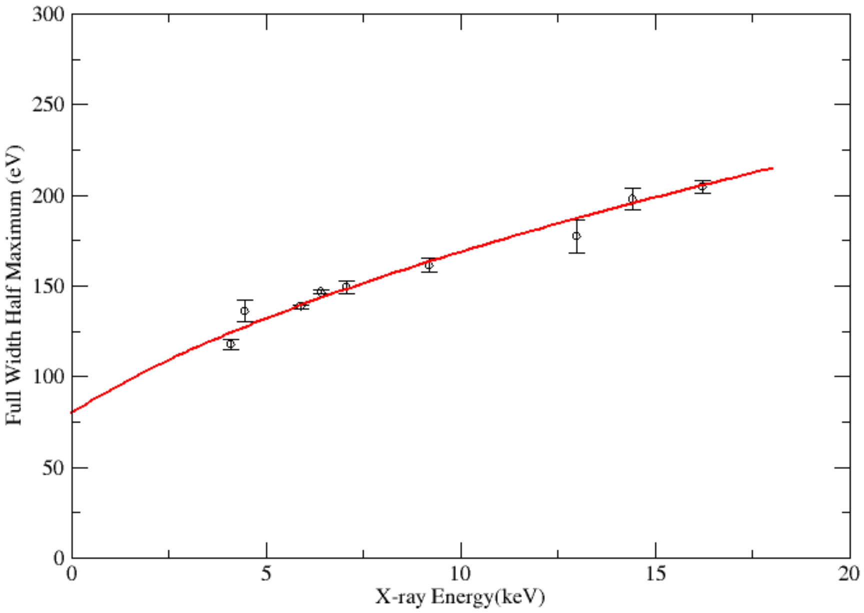
Silicon Drift Detector





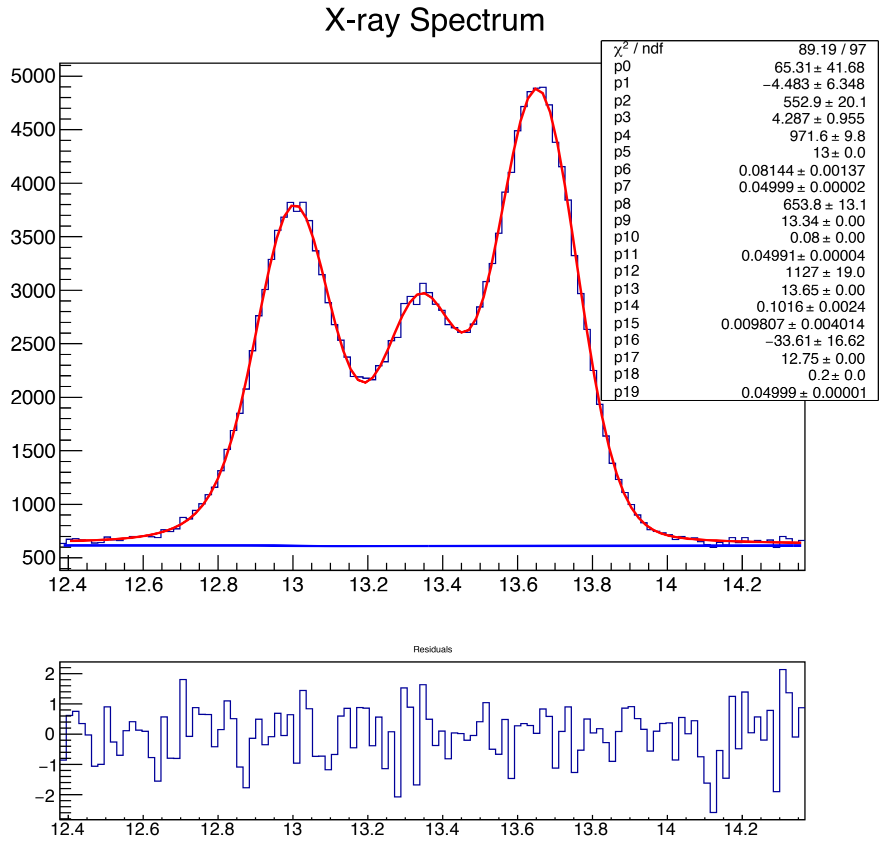
Silicon Drift Detector Resolution

- The resolution is determined by the Fano limit and detector noise.
- The fit used a Fano factor of 0.11 and a 33 eV (sigma) noise added in quadrature.



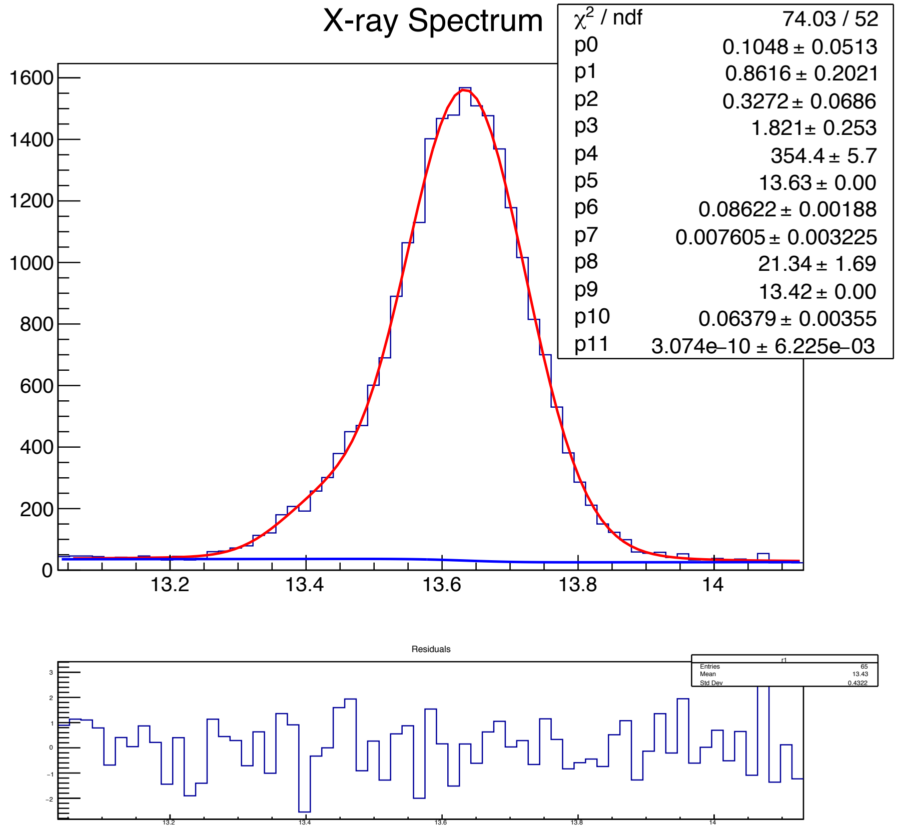
X-ray Spectra Fit to the Voigt Profile

- The Voigt profile is a convolution of the Lorentzian distribution with a Gaussian.
- Depleted uranium has a Thorium x-ray, 13 keV, protactinium at 13.34 keV, and uranium at 13.65 keV

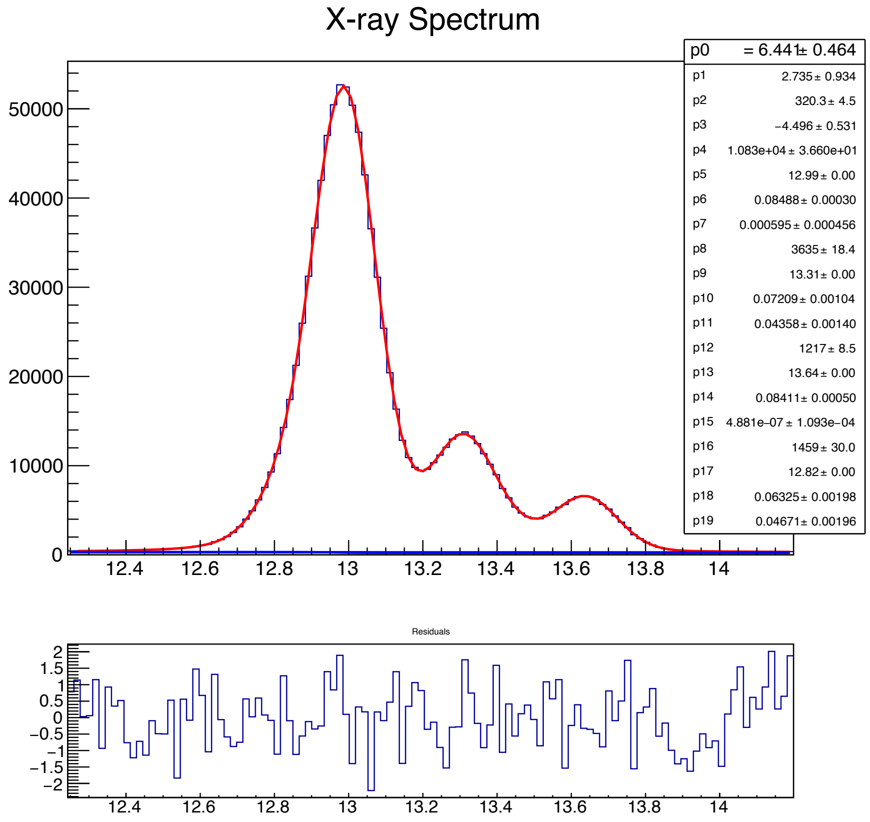


χ^2 / ndf	89.19 / 97
p0	65.31 ± 41.68
p1	−4.483 ± 6.348
p2	552.9 ± 20.1
p3	4.287 ± 0.955
p4	971.6 ± 9.8
p5	13 ± 0.0
p6	0.08144 ± 0.00137
p7	0.04999 ± 0.00002
p8	653.8 ± 13.1
p9	13.34 ± 0.00
p10	0.08 ± 0.00
p11	0.04991 ± 0.00004
p12	1127 ± 19.0
p13	13.65 ± 0.00
p14	0.1016 ± 0.0024
p15	0.009807 ± 0.004014
p16	−33.61 ± 16.62
p17	12.75 ± 0.00
p18	0.2 ± 0.0
p19	0.04999 ± 0.00001

Comparison of Pu-239, U-235 L X-rays



Pu-239



HEU

Alpha Counting

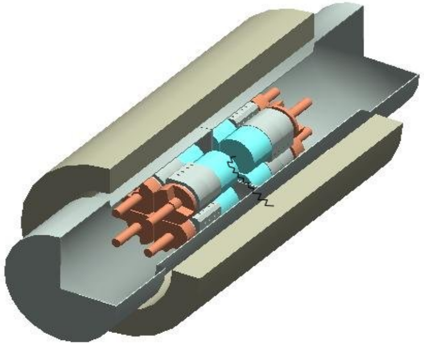
- Detectors are silicon surface barrier detectors, Frisch grid ionization chamber, and 2-Pi gas proportional counters.
- The energy resolution is typically limited by sample preparation. The best deposits require:
  - Clean chemical separation from contaminants.
  - Perchloric acid fuming to remove organic contaminants
  - Electrodepositing on platinum
  - Flaming to remove any other residues

Detector Structure

- **Clover Structure (2 sets of 4 crystals)**
  - Each crystal ~55 mm diam x 70 mm long
  - Quantify gamma rays in cascades and utilize coincidence counting methods
  - Preserve high resolution while attaining a large volume and detector face area
    - Environmental samples can be quite large
    - Accommodate a wide range of sample sizes and shapes

Shielding / Structure / Background

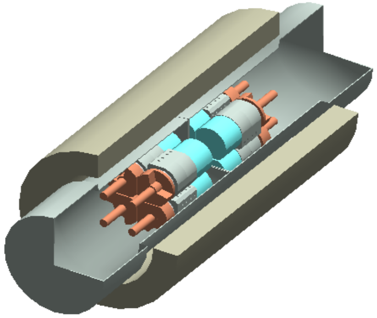
- **Compton Suppressed**
  - Reduce cosmic ray induced background
    - – Lab sits at ~7500' elevation
  - Reduce Compton continuum



- **Low Background**
  - Low-Z selected materials stand and structure
    - – Stand fabricated entirely from plastics
  - Special shielded room
    - – Entire room is the shield, 12” thick pre-WWII steel

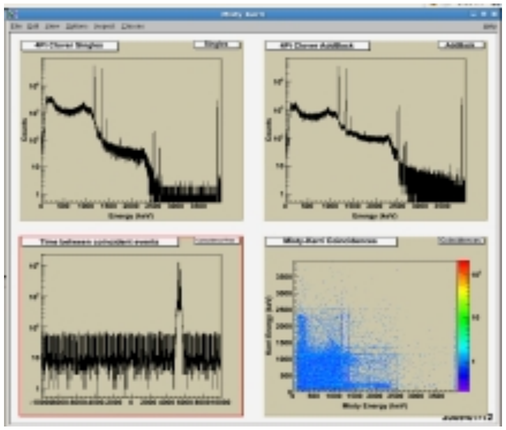
Compton suppression

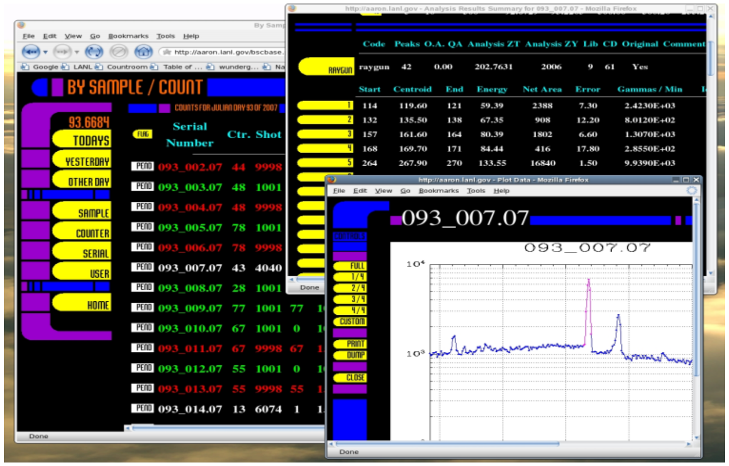
- **Modeled using GEANT prior to purchase**
- **Custom made NaI (Tl) detector**
  - 2” thick walls, 16” long
  - OFHC Copper outer jacket
  - Thin copper and plastic inner jacket
  - Reduced background ~ 10x



Data Acquisition System

- **Digital Spectral Data Processing**
  - Each channel digitized and time-tagged as stored data.
    - Commercial hardware, custom software
  - Fully integrated with Web-based visualization system
    - Readout singles, sum-of-singles, full addback modes





	Serial Number	Ctr.	Shot	
PEND	093_002.07	44	9998	
PEND	093_003.07	48	1001	
PEND	093_004.07	48	9998	
PEND	093_005.07	78	1001	
PEND	093_006.07	78	9998	
PEND	093_007.07	43	4040	
PEND	093_008.07	28	1001	
PEND	093_009.07	77	1001	77
PEND	093_010.07	67	1001	0
PEND	093_011.07	67	9998	67
PEND	093_012.07	55	1001	0
PEND	093_013.07	55	9998	55
PEND	093_014.07	13	6074	1

Code	Peaks	O.A. QA	Analysis ZT	Analysis ZY	Lib	CD	Original	Comment
raygun	42	0.00	202.7631	2006	9	61	Yes	

	Start	Centroid	End	Energy	Net Area	Error	Gammas / Min
1	114	119.60	121	59.39	2388	7.30	2.4230E+03
2	132	135.50	138	67.35	908	12.20	8.0120E+02
3	157	161.60	164	80.39	1802	6.60	1.3070E+03
4	168	169.70	171	84.44	416	17.80	2.8550E+02
5	264	267.90	270	133.55	16840	1.50	9.9390E+03

Data Processing

- **All data is captured event-by-event, the pulse height determined, and stored with a 13 ns precision time tag.**
- **The Compton suppressor used to selectively veto events.**
- **Data is processed post-run in one or more ways**
  - Any individual single HPGe crystal, with or without the Compton suppression active
  - **Singles** mode is the sum of all HPGe detector events, with **no regard to timing,** with veto active (Singles with veto) or inactive (Singles)
  - **Addback** mode is where HPGe events **coincident in time are added together** to make a single event, with veto active (Addback with veto) or without veto active (Addback)

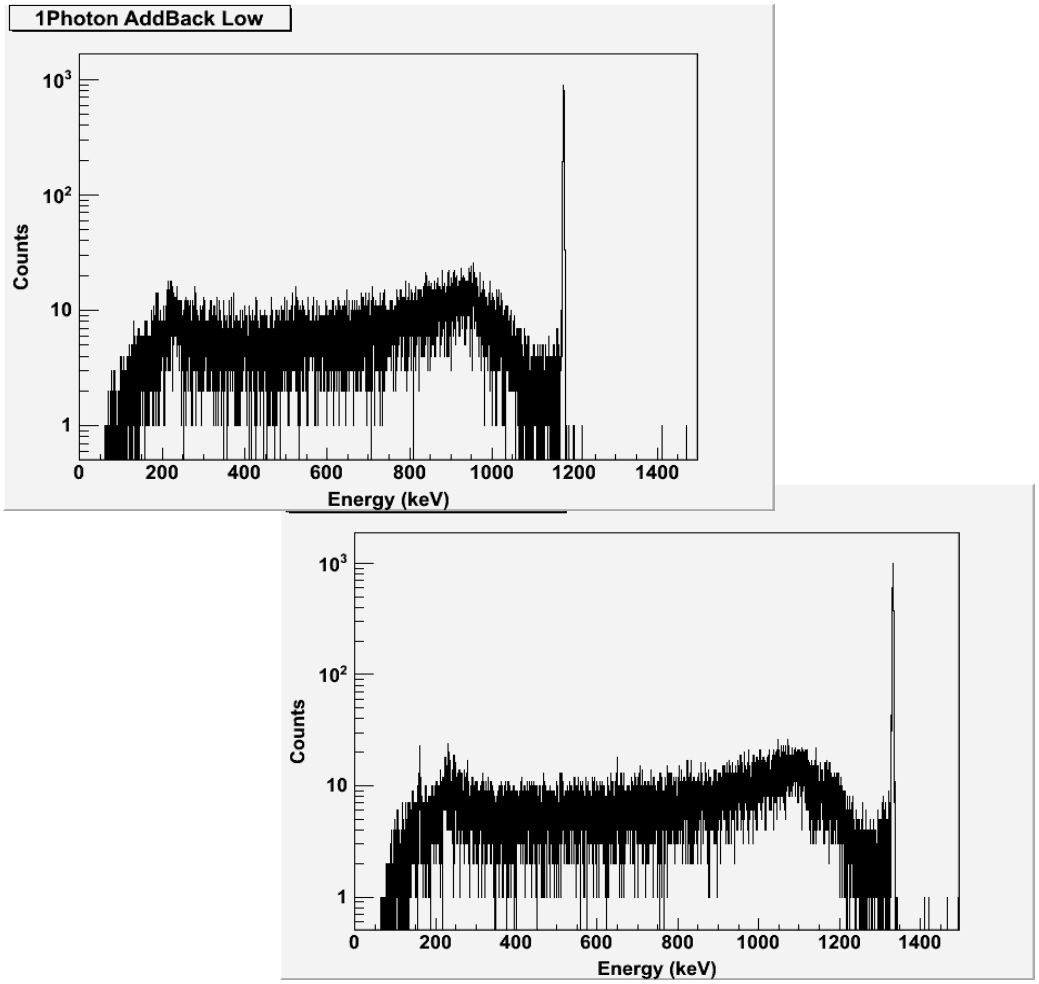
Photo-Peak Efficiencies

- Gamma ray cascades complicate photo-peak efficiencies in high geometry detectors. The rate under a photo peak for a 2 gamma ray cascade is described by the following:

$$R_1^{PhotoPeak} = \varepsilon_1^{PhotoPeak} f_1 * \left(1 - \varepsilon_2^{Total} f_2\right) * Activity$$

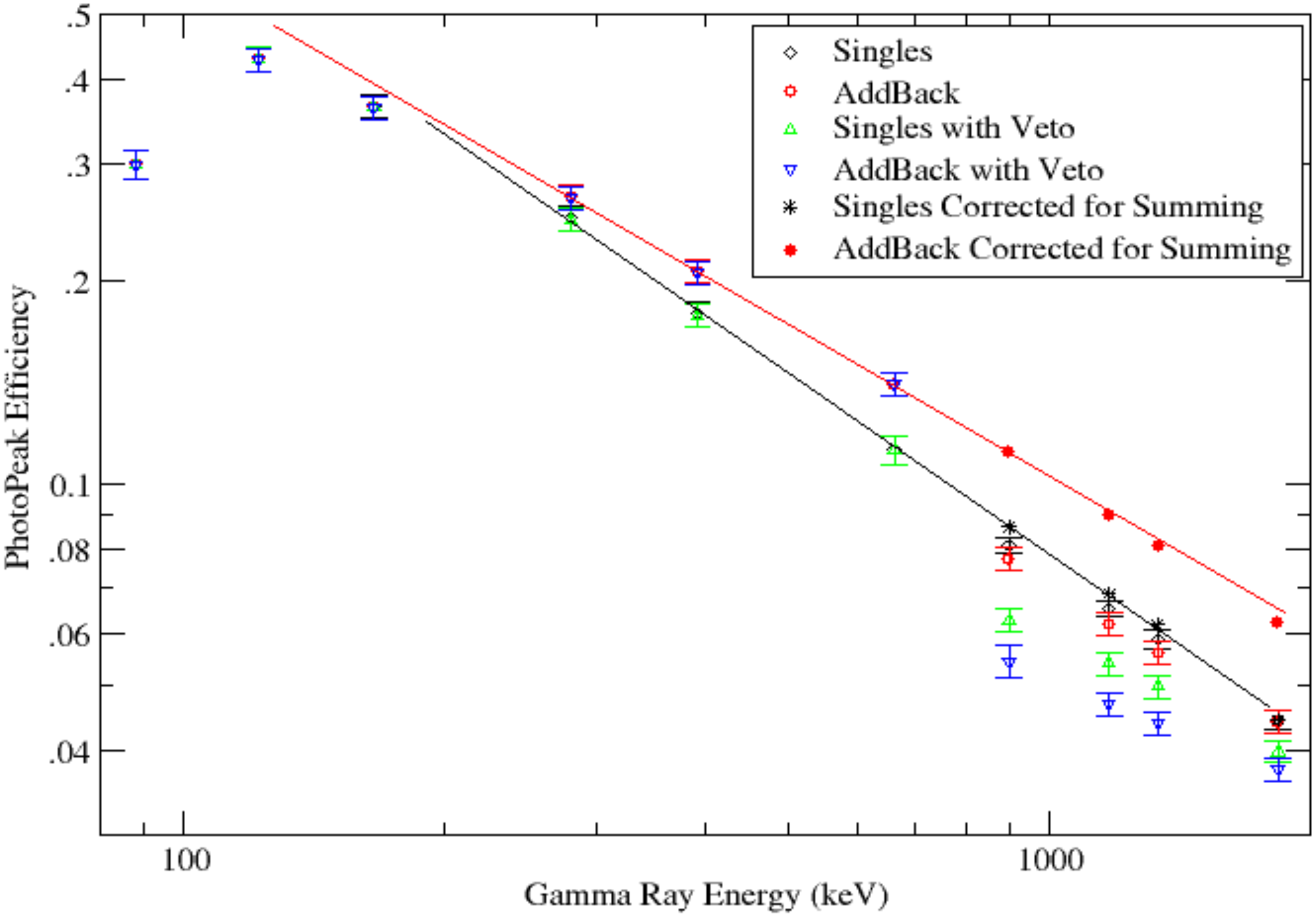
- The decay scheme must be known to calculate the apparent photo-peak efficiency. The true photo-peak efficiencies and the total efficiencies must be known as a function of gamma ray energy.
- The total efficiency for the NaI veto has yet to be determined.

Single Photon Spectra from a 2 Gamma Ray Cascade

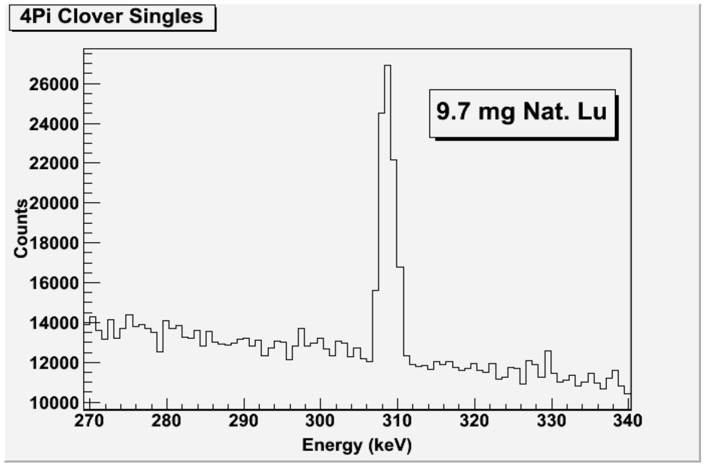


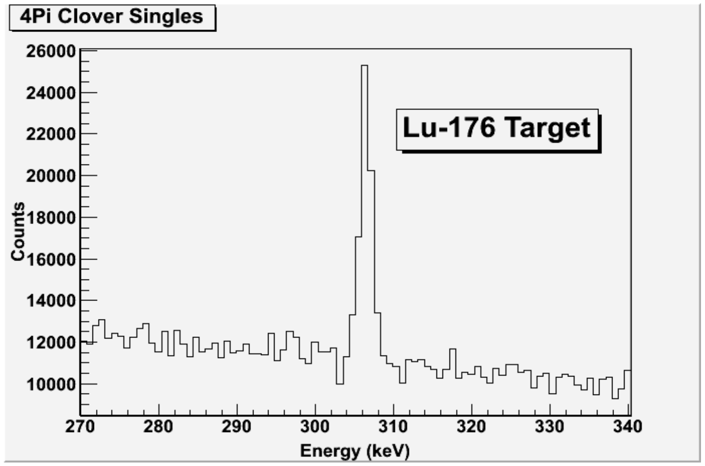
- If a single crystal has an event with full energy from a single photon, the coincident events in the remaining crystals are histogrammed.
- This is one of the advantages of list mode data.
- Peak to total efficiencies can then be determined from these spectra.
- Example is Co-60.

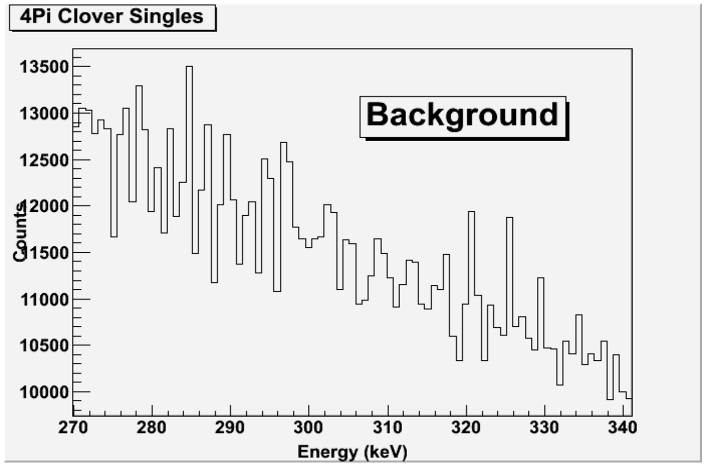
Clover Efficiencies



Lu-176 Determined Relative to Natural Lutetium







Mass Determination

- Lu-176 target 7 mm diameter 35683 counts ± 433 in 8000 minutes
- 9.7 mg natural Lu foil diameter is 6.35 millimeter
- foil thickness is 30.6 milligrams per square centimeter
- Area under the 306.8 keV peak 43438 counts ± 720 in 8000 minutes
- 0.216 counts per minute per microgram Lu-176
- Target mass is 209 micrograms Lu-176